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Title: Iridium an Important Neutron Flux Monitor

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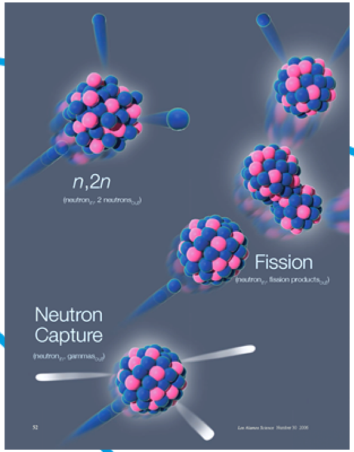
Abstract

Iridium has been used for decades at Los Alamos as a fluence monitor in underground nuclear testing. Iridium can be used to monitor three neutron spectral groups at once, thermal/epithermal, 14 MeV, and fission spectrum (>2 MeV). The nuclear reaction Ir-193(n,n')Ir-193m has not had a model independent cross section, until recently. The first measurement of the cross section was made last fall at the Berkeley HFNG.



Iridium an Important Neutron Flux Monitor

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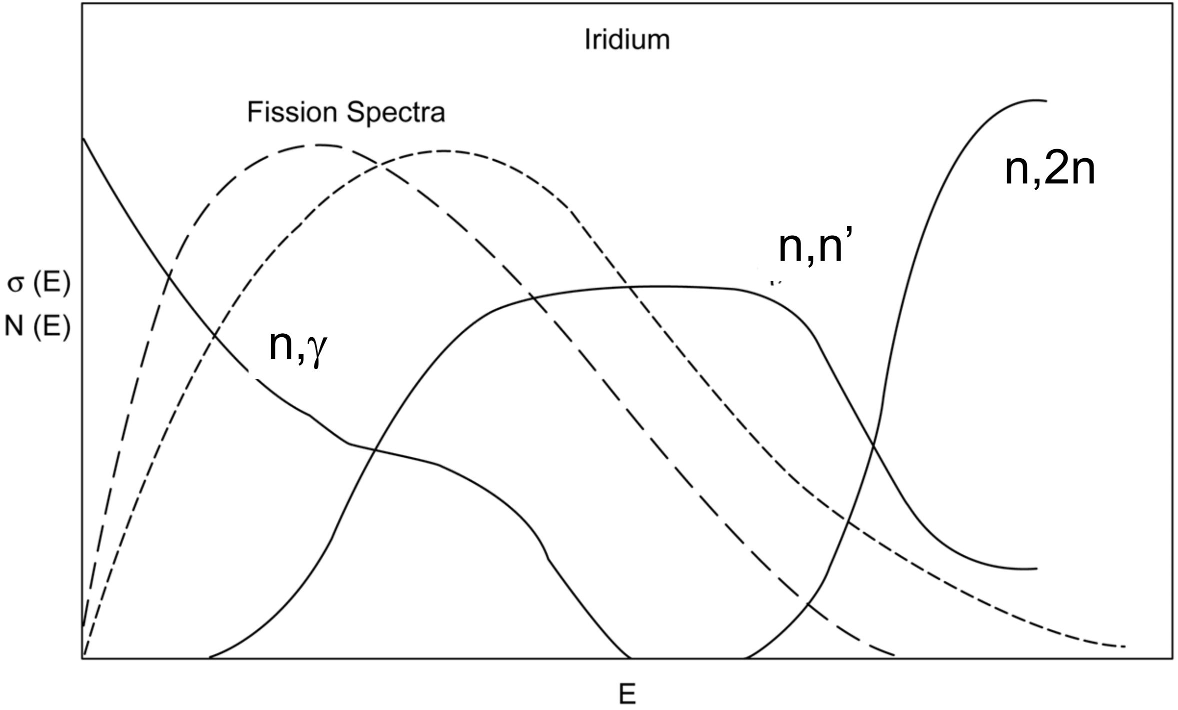
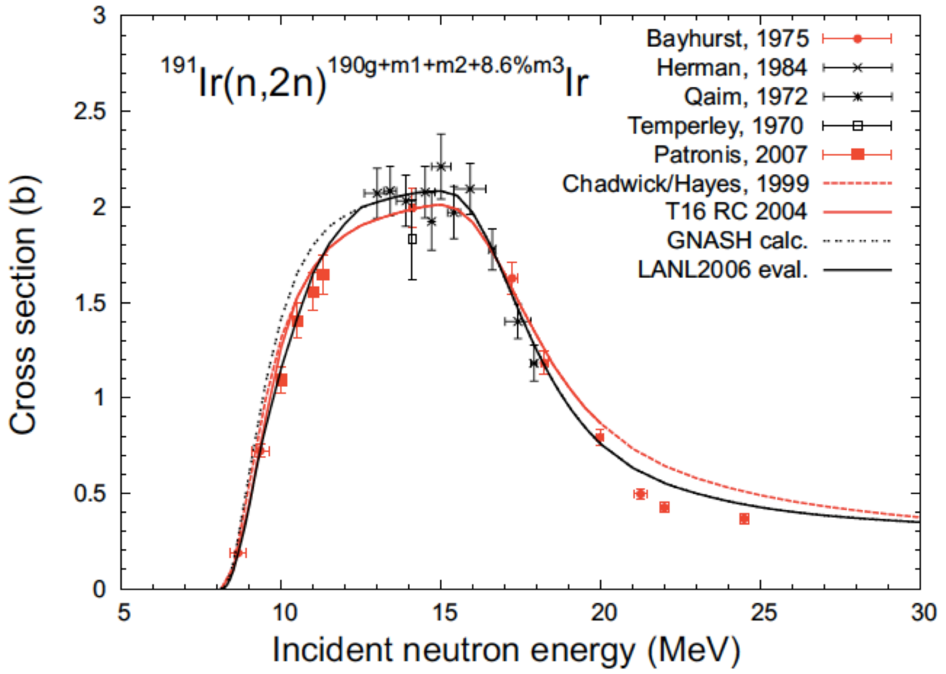
Iridium Nuclear Reactions Sample Three Energy Groups

- Iridium is used as a Rad Chem detector for measurement of neutron fluence. Accurate data on neutron-induced cross sections for the two stable iridium isotopes (^{191}Ir and ^{193}Ir) are of importance to stockpile stewardship.
- Iridium activation may be important in post-detonation nuclear forensics.



Nuclear Data: $^{191}Ir(n,2n)^{190}Ir$ Cross Section

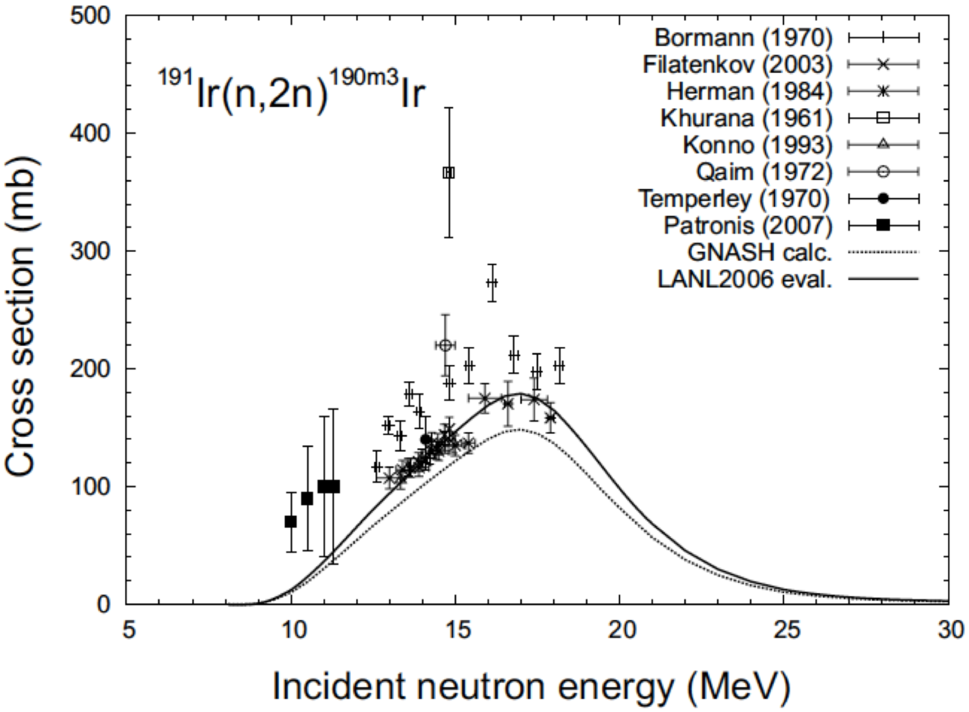
The new evaluation (LANL2006, solid line) for $^{191}Ir(n,2n)^{190g+m1+m2+8.6\%m3}Ir$ is compared with experimental data and with previous evaluations and calculations. This combination of g+m1+m2+8.6%m3 is used because it represents what is typically measured in an activation measurement. The black dashed line represents the GNASH result, which was adopted for the latest LANL2006 evaluation (black solid line) at all energies except below 12 MeV where the evaluation was adjusted to better agree with the Bayhurst data points and with integral information from critical assembly reaction rates. As a comparison, the 1999 calculation by Chadwick and Hayes (dashed red line) and the T16 RC 2004 evaluation (solid red line) are also shown.



Nuclear Data Sheets 108 (2007) 2716–2741

Nuclear Data: ^{191}Ir(n,2n)^{190m3}Ir Cross Section

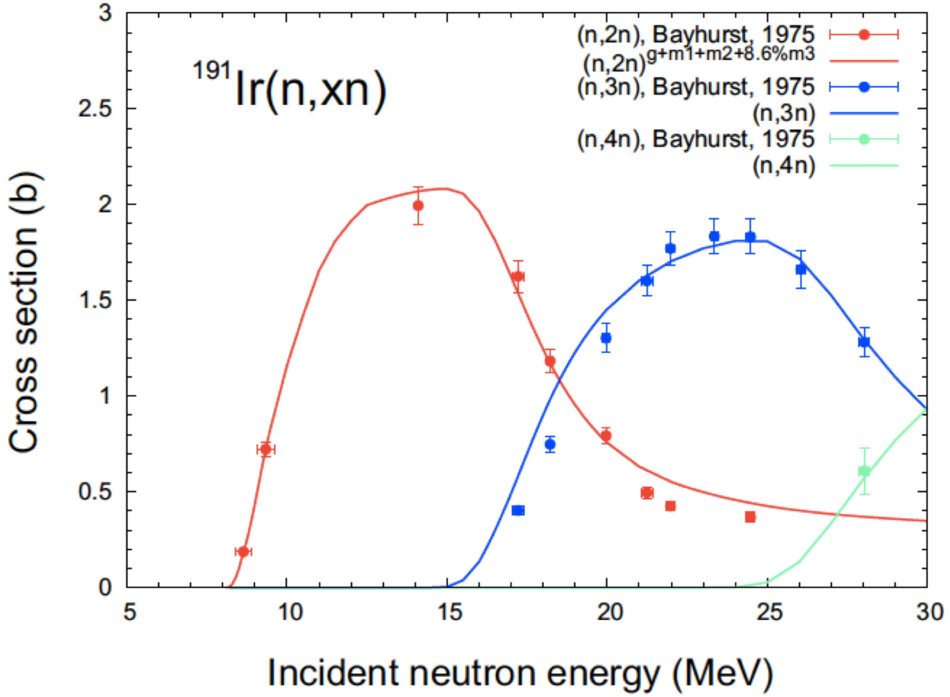
- The evaluated 191Ir(n, 2n) excitation function to the m3 isomer in 190Ir is compared with experimental data.
- The new evaluation (solid line) was obtained by scaling the GNASH calculated result by a factor of 1.205, in order to better agree with the bulk of experimental data.
- The isomer has a 3.2 hour half-life.



Nuclear Data Sheets 108 (2007) 2716–2741

Nuclear Data: ^{191}Ir(n,2n)^{190,189,188}Ir Cross Sections

- Evaluated LANL2006 ^{191}Ir (n, 2n), (n, 3n) and (n, 4n) cross sections compared with measurements.
- These reactions can be important in inertial confinement fusion experiments.
- 190Ir, 189Ir, and 188Ir have half-lives of 11.8, 13.2, and 1.72 days, respectively.



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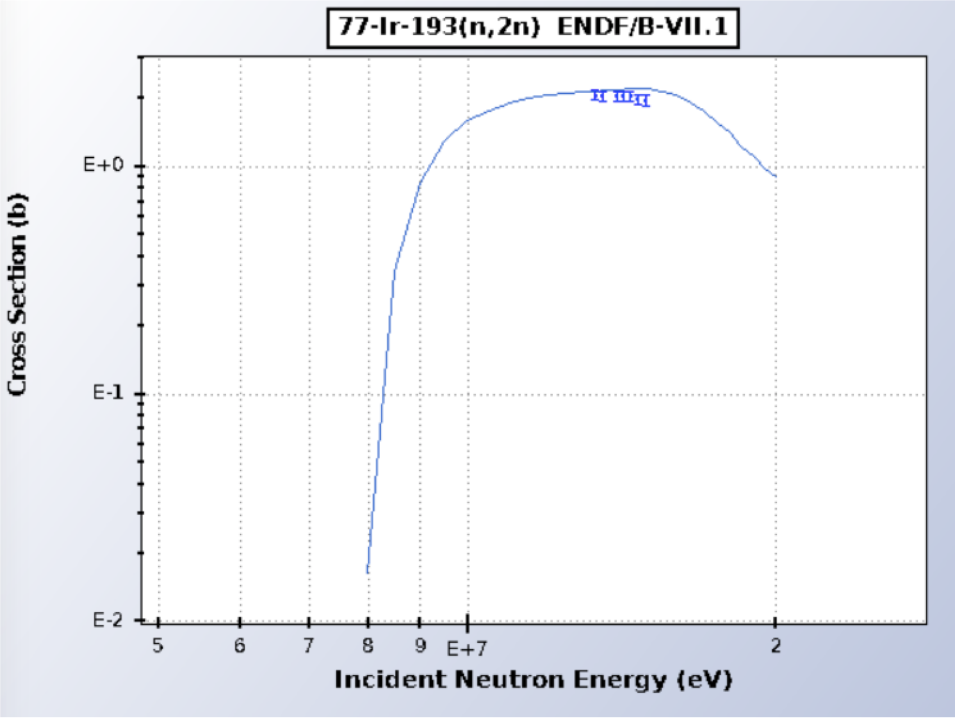
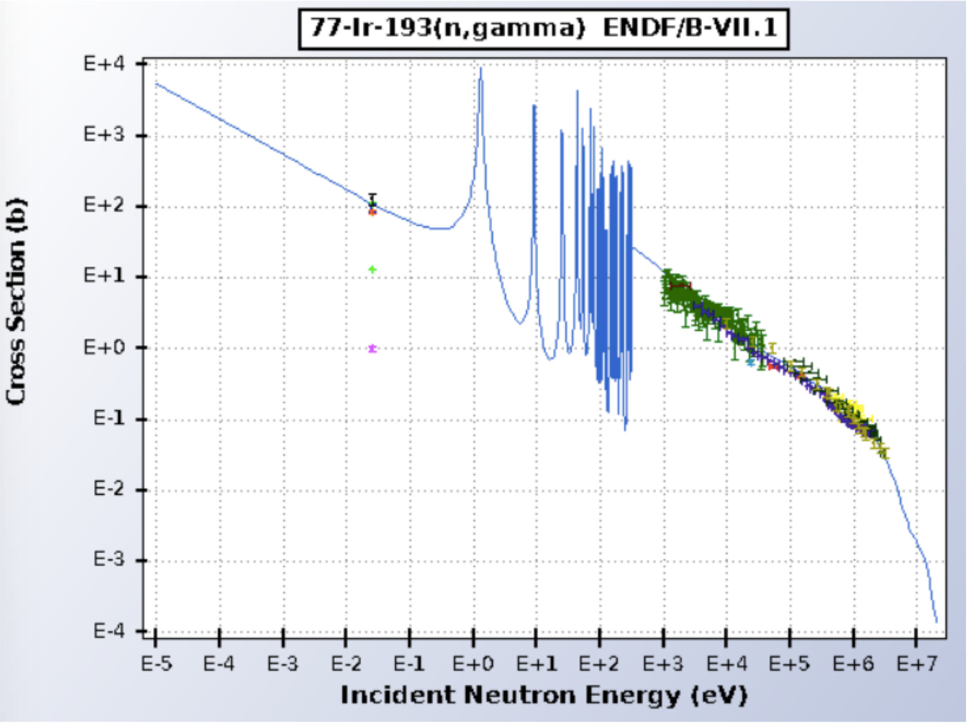
Nuclear Data: $^{193}Ir(n,2n)^{192}Ir$ Cross Section



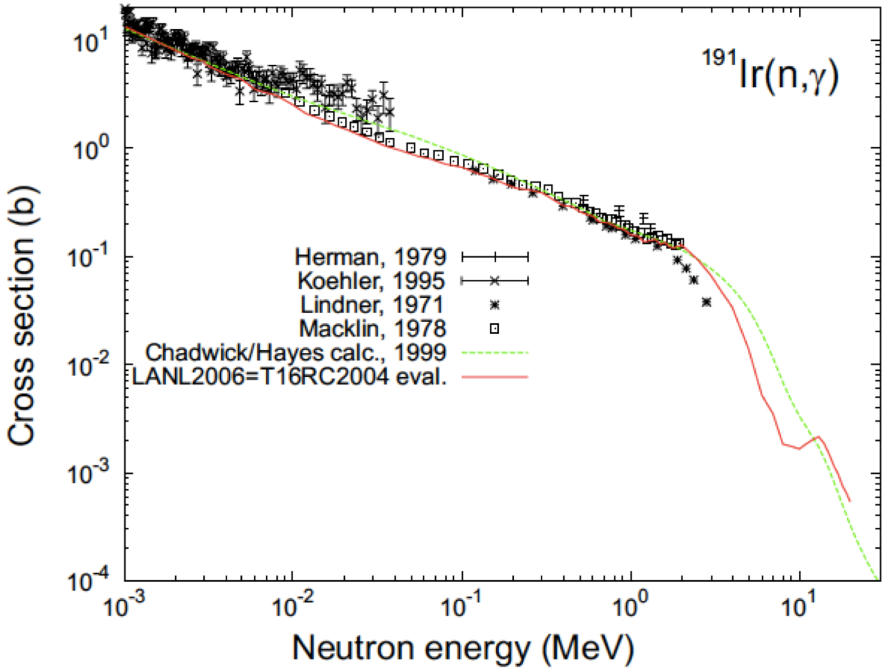
Nuclear Data: $^{193}Ir(n,\gamma)^{194}Ir$ Cross Section

- Experimental data is lacking in the resonance region.
- Measurements at DANCE are proposed for the next beam cycle.
- ^{194g}Ir has a 19.3 hour half-life and abundant gamma rays.
- ^{194m}Ir has a 171 day half-life.



Nuclear Data: $^{191}Ir(n,\gamma)^{192}Ir$ Cross Section

- The ^{191}Ir (n,γ) cross section in the present LANL2006, which is identical to the the T16 RC 2004 evaluation, compared with measurements.
- The evaluation was chosen in part to optimize agreement with the integral critical assembly reaction rate data as described in this paper.
- ^{192}Ir has 73.83 day half-life.



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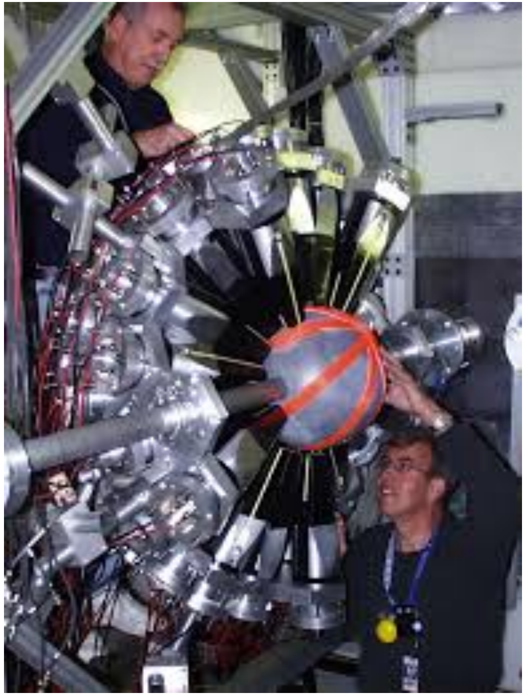
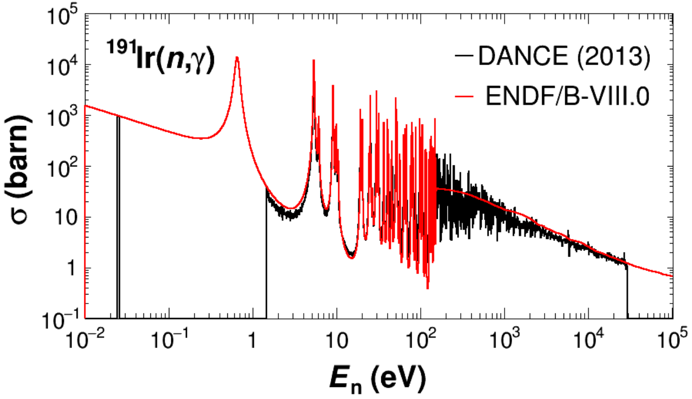
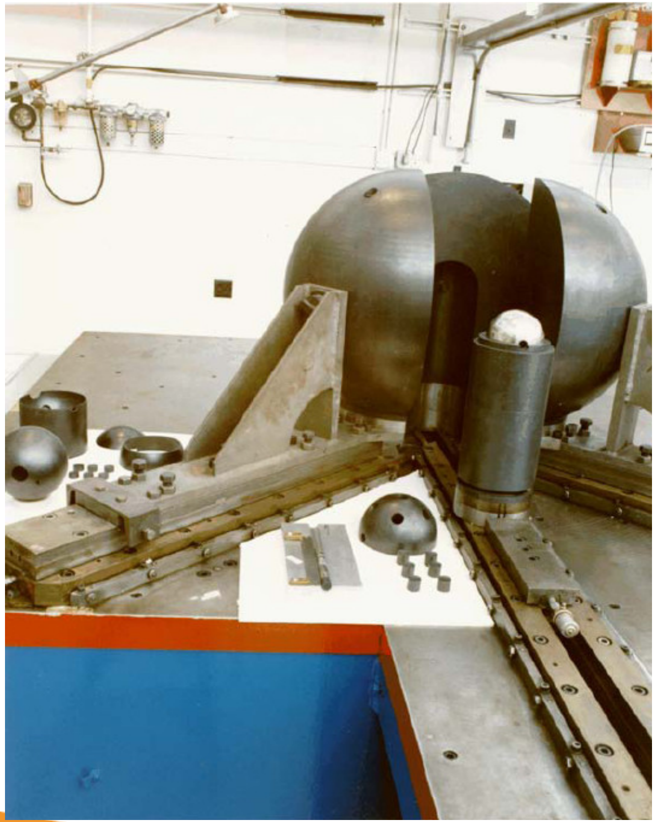
Differential Measurements of Neutron-Induced Cross Sections from Stable Iridium Isotopes

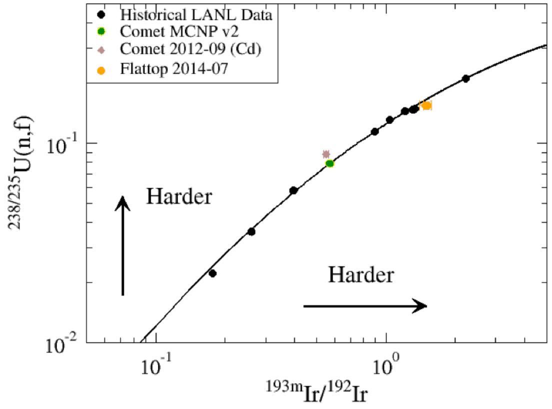




- Currently the uncertainty of the ENDF cross sections is above 10-15% for E_n > 100 keV.
- New measurements with improved DAQ are planned, they will scan the energy range from E_{th} to 1 MeV.
- Measurements with targets with different ^{191}Ir/^{193}Ir ratio to obtain the (n,γ) cross section from each isotope.

Integral Measurements of Neutron-Induced Cross Sections from Stable Iridium Isotopes



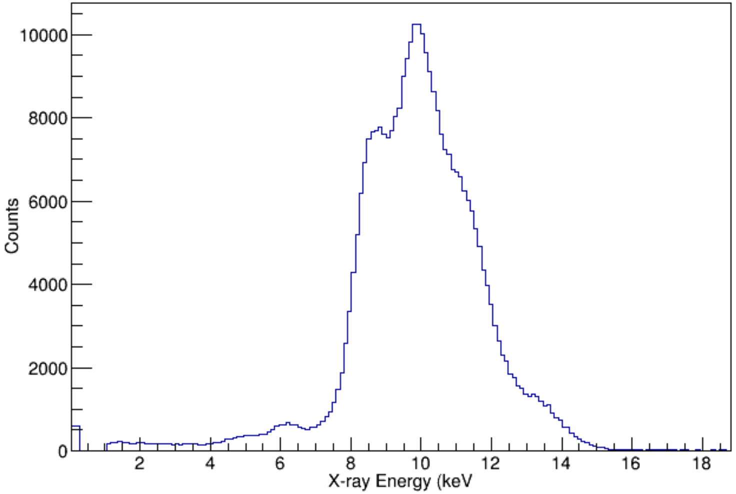


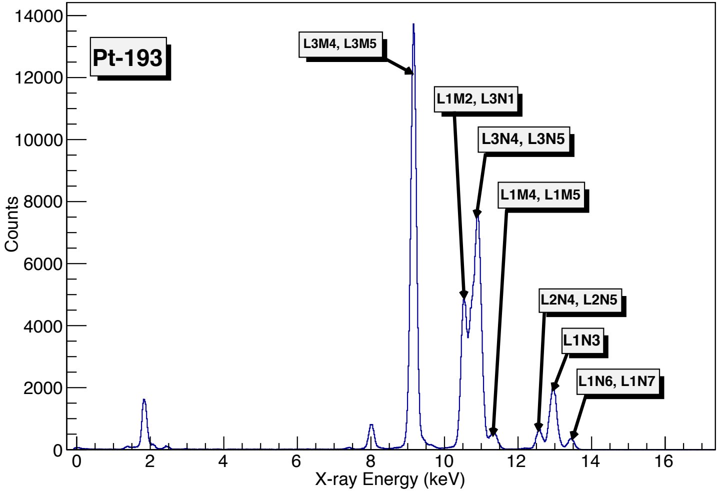
- The ratio of ^{193m}Ir (produced by fission spectrum neutron) to ^{192}Ir (produced by neutron capture) is a metric of spectral hardness.
- Integral measurements of this ratio, recently at NCERC, in Los Alamos critical assemblies have verified the historical data.
- Accurate ^{192}Ir(n, γ) cross sections are needed to correctly simulate nuclear detonations.

Assay of ^{193m}Ir

- ^{193m}Ir decays by an M4 transition and is highly converted, the abundance of the 80.3 keV gamma ray is too small to be practical.
- The abundance of the K x-rays are also small. Therefore the assay is typically done by measuring the L x-rays.
- The L x-rays have energies between 9 and 10 keV. Self-attenuation requires that thin deposits be prepared. This necessitates chemical dissolution and precipitation of cesium hexachloro-iridate.
- Measurements in critical assembly experiments use soluble potassium hexachloroiridate as a target.
- After irradiation, the potassium is chemically separated away and cesium hexachloroiridate is precipitated out.
- An 11 mm diameter deposit of 10 to 30 mg is formed on a filter paper. The deposit was historically counted using a argon gas proportional counter with a beryllium window.
- The gas proportional counter could not resolve iridium x-rays from osmium and platinum x-rays from the other isotopes of iridium. A deconvolution method was employed.
- Los Alamos is developing new methodology employing a silicon drift detector to improve the assay method.

New Technology: Silicon Drift Detector Improves the Measurement of ^{193m}Ir



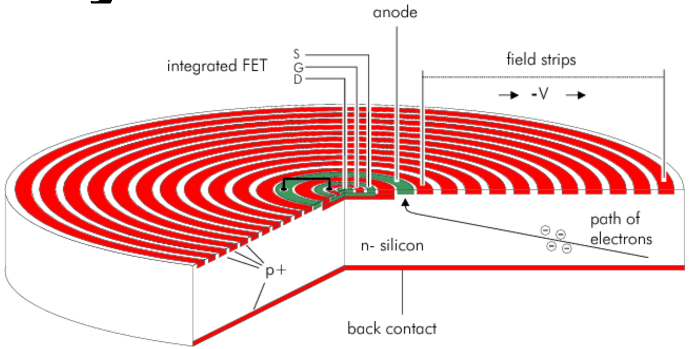


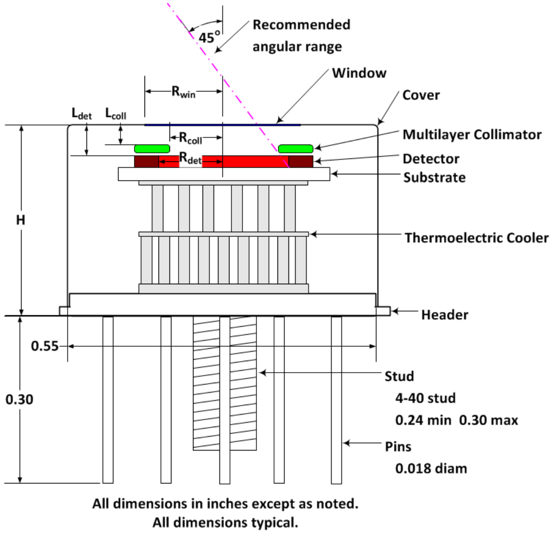
The figure shows Ir L x-rays from electron Capture in the ^{193}Pt standard.
The gas proportional counter used during underground testing cannot resolve individual X-rays. The resolution is about 11 percent.

The Silicon Drift Detector, SDD, has a Resolution of about 0.15 percent. It is capable of resolving Pt, Ir, and Os L x-rays.

A Silicon Drift Detector for ^{193m}Ir L X-rays

- Superior performance over a gas proportional counter.
- The charge is directed to an internal FET.
- This lowers noise pickup.





- The SDD is thermoelectrically cooled via Peltier junctions.
- The resolution that can be obtained is 123 eV on manganese K X-rays from 55Fe electron capture.

X-ray Line Shape

The Voigt profile is a convolution of a Gaussian profile with a Lorentzian profile.

$$V(x;\sigma,\gamma) = \int_{-\infty}^{\infty} G(x';\sigma)L(x-x';\gamma)\,dx',$$

where x is the shift from the line center, $G(x;\sigma)$ is the centered Gaussian profile:

$$G(x;\sigma) \equiv \frac{e^{-x^2/(2\sigma^2)}}{\sigma\sqrt{2\pi}},$$

and $L(x;\gamma)$ is the centered Lorentzian profile:

$$L(x;\gamma) \equiv \frac{\gamma}{\pi(x^2+\gamma^2)}.$$

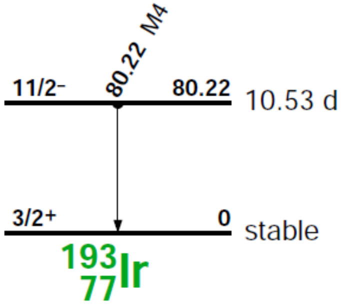
The defining integral can be evaluated as:

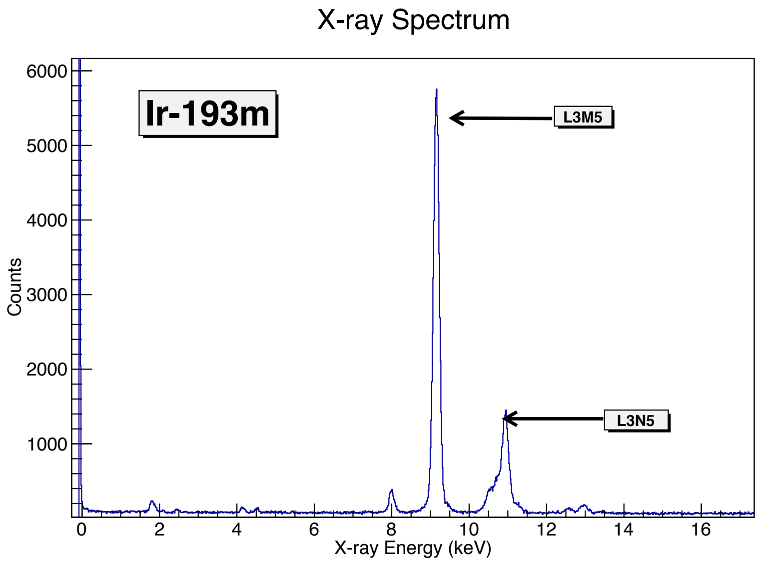
$$V(x;\sigma,\gamma) = \frac{\mathrm{Re}[w(z)]}{\sigma\sqrt{2\pi}},$$

where Re[$w(z)$] is the real part of the Faddeeva function evaluated for

$$z = \frac{x+i\gamma}{\sigma\sqrt{2}}.$$

^{193m}Ir Produced in Flattop

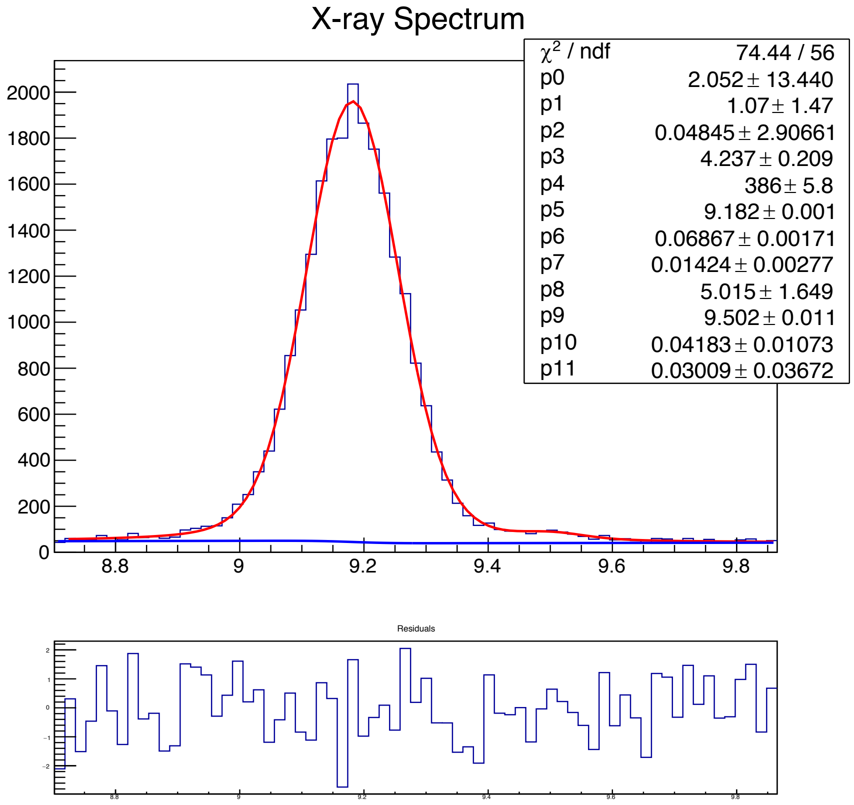




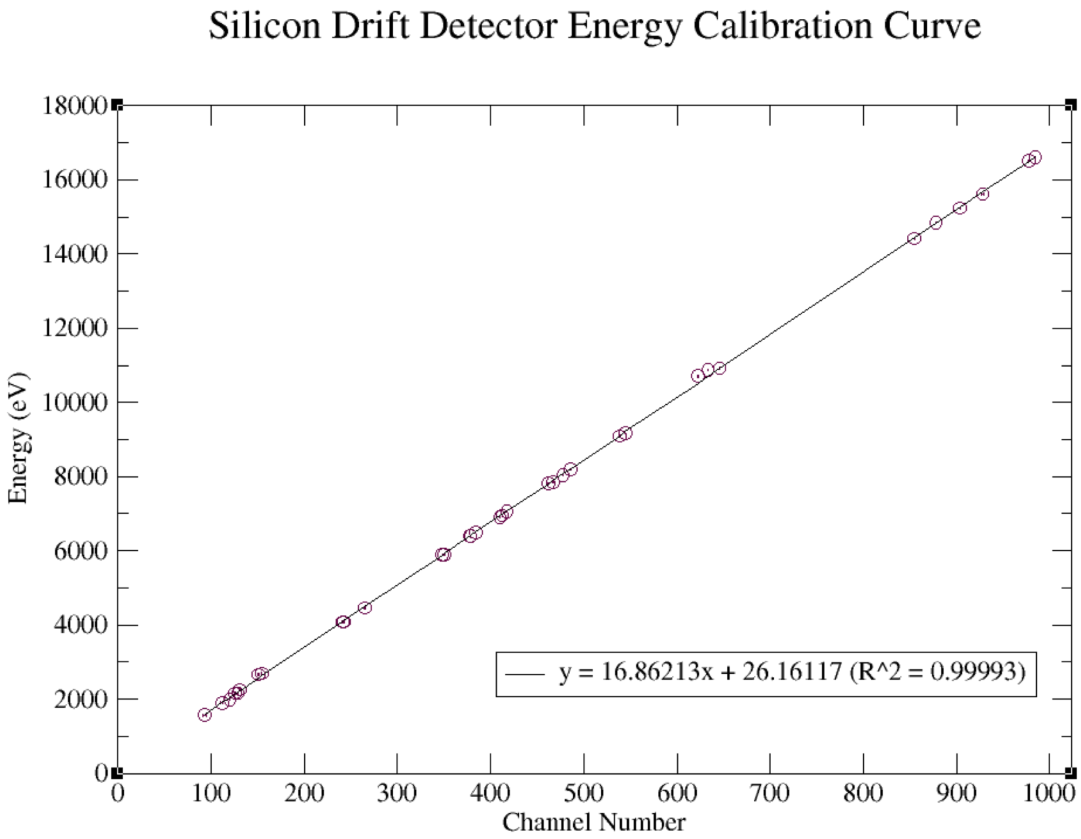
- The M4 transition favors high angular momentum electrons in the L-shell.
- This may be the cause of the factor of two discrepancy in the atom-scale.

Voigt Profile Fit to ^{193m}Ir L X-ray

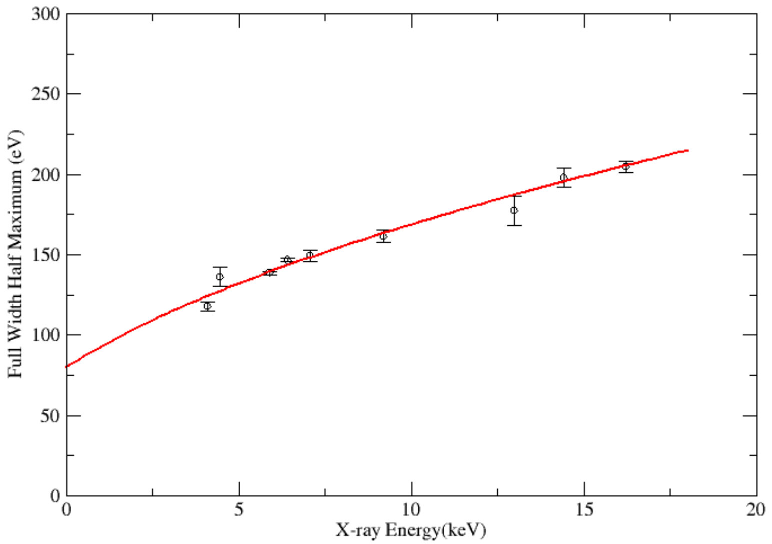
- The Voigt profile provides a nearly perfect fit to the SDD L x-ray.
- The Platinum L x-ray was also fit at 9.5 keV.
- Only two points out of sixty are above 2 sigma.



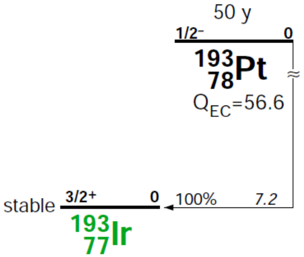
SDD linearity

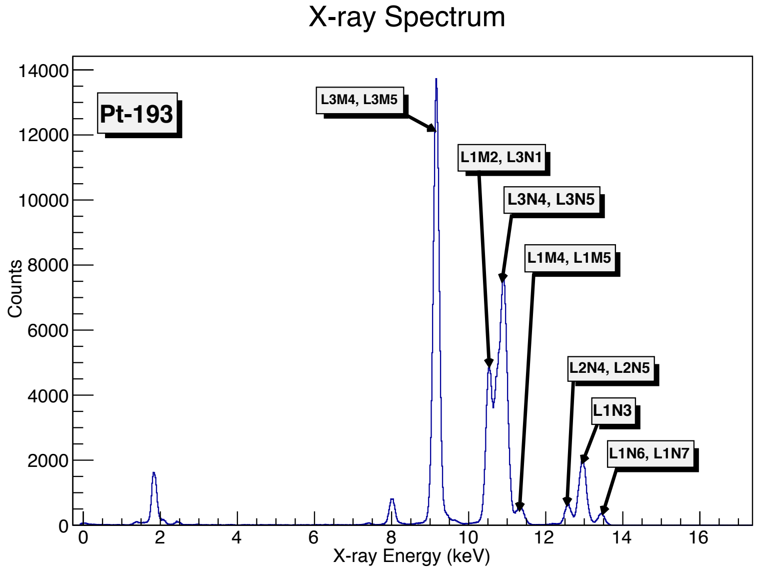


SDD Resolution



^{193}Pt Calibration Source





- In the course of calibrating our detectors, we discovered that the half-life of ^{193}Pt is incorrect. It is actually 400 years.
- It is still a first forbidden transition. Therefore low angular momentum electrons can be captured from the L-shell.

Iridium Inelastic Scatter Provides a Measure of Fission Neutron Flux

As was done in the past at Los Alamos, we determine this detector efficiency through comparisons of the absolute magnitude of our simulation code predictions (e.g. MCNP using our nuclear data), with the critical assembly measurements, and we conclude here that the detector efficiency is about 0.5 - i.e., measured ^{193m}Ir values need to be multiplied by a factor of 2.0 before comparing with the simulations. This factor of 2.0 is identical to that applied by LANL experimentalists [25] when reporting their ^{193}Ir(n, n') cross sections in their 1970s Bayhurst experiment in the Physical Review (and shown in our Fig. 14), as reported to us by Gilmore and by Barr [59].

M.B. Chadwick et al., Nuclear Data Sheets 108 (2007) 2716ñ2741

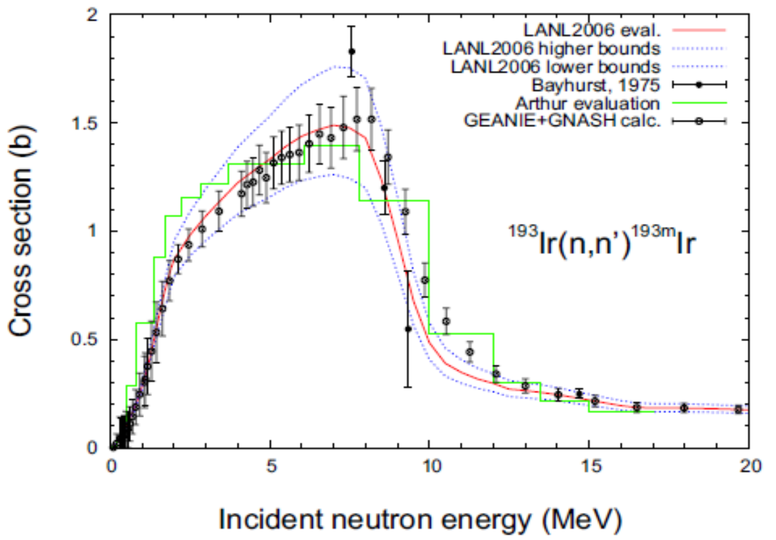
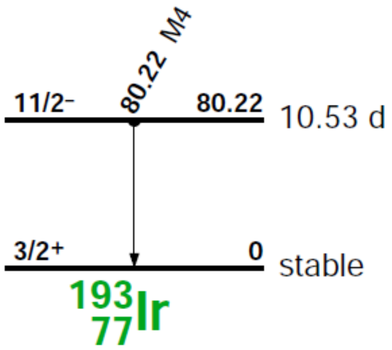
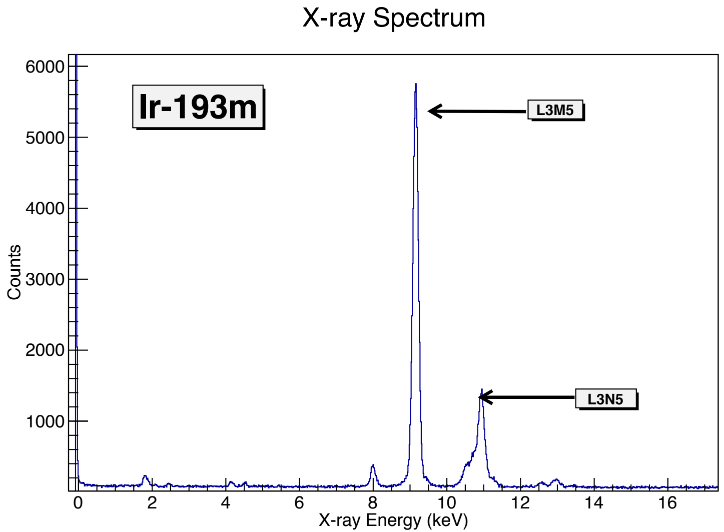


FIG. 14: Total isomer production cross section through the reaction ^{193}Ir (n,n') ^{193m}Ir. The new LANL2006 evaluation is shown by the bold red line, and the blue dotted lines show the uncertainty bounds that we estimate. The experimental results from the GEANIE+GNASH analysis, shown as black circles, was used for the earlier T16_RC_2004 evaluation. The histogram given as the green line describes the old multigroup Arthur evaluation.

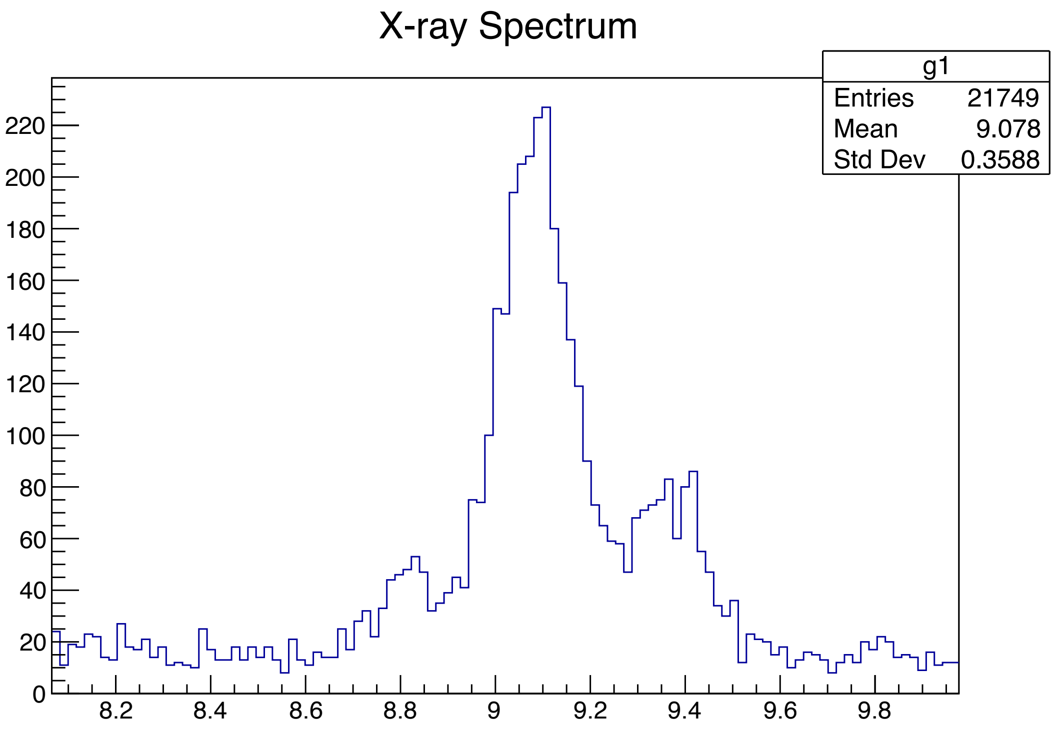
^{193m}Ir Produced in Flattop





- The M4 transition favors high angular momentum electrons in the L-shell.
- This may be the cause of the factor of two discrepancy in the atom-scale.

160 Day Old Iridium Irradiated in Flattop



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Iridium Activation Applied to Critical Assembly Spectra

- The evaluated cross sections differ from the historical data.
- We hope to be able to more accurately establish the modern to historical atom ratio for ^{193m}Ir.
- The Chadwick evaluation uses approximately a factor of two.
- Other factors have been used 2.379 to 2.75.
- We hope to better define the modern to historical atom ratio.

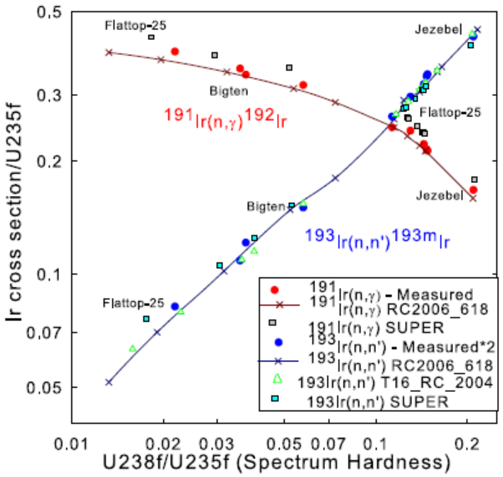
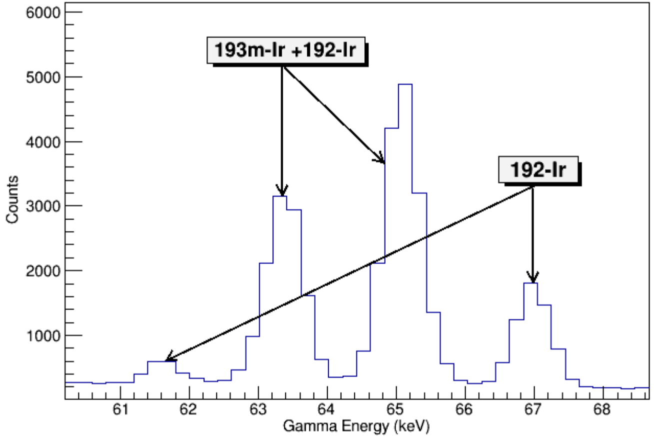
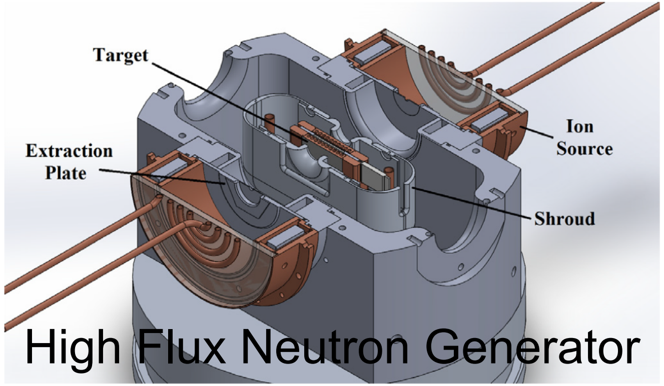


FIG. 18: Critical assembly reaction rate measurements, compared with simulations, for the ^{191}Ir $(n,\gamma)^{190}$Ir cross section and the ^{193}Ir$(n,n')^{193m}$Ir cross section. The x-axis shows the spectral index, which is a measure of the hardness of the neutron spectrum. The new LANL2006 data are referred to as RC2006_618 in this figure. The SUPER data refer to the old evaluation by Arthur, and overpredict the (n,n') measurements in the soft spectrum regions on the left hand side of the figure. The LANL2006 (n,n') evaluation performs better here because of its smaller cross section rise from threshold below a few MeV.

First Measurement of the Activation Cross Section for ^{193}Ir(n,n')^{193m}Ir and ^{191}Ir(n, γ)^{192}Ir at 2.7MeV Neutron Energy using the UC Berkeley High Flux Neutron Generator

- The ^{193m}Ir 10.53 day isomer is a valuable radiochemical diagnostic.
- The Population of the isomer has only been measured at En>7.5 MeV or indirectly.
- We measured the ^{193}Ir(n,n')^{193m}Ir and ^{191}Ir(n, γ)^{192}Ir cross sections relative to the well known ^{nat}Ni(n,p)^{58}Co and ^{nat}In(n,n')^{115m}In reactions.



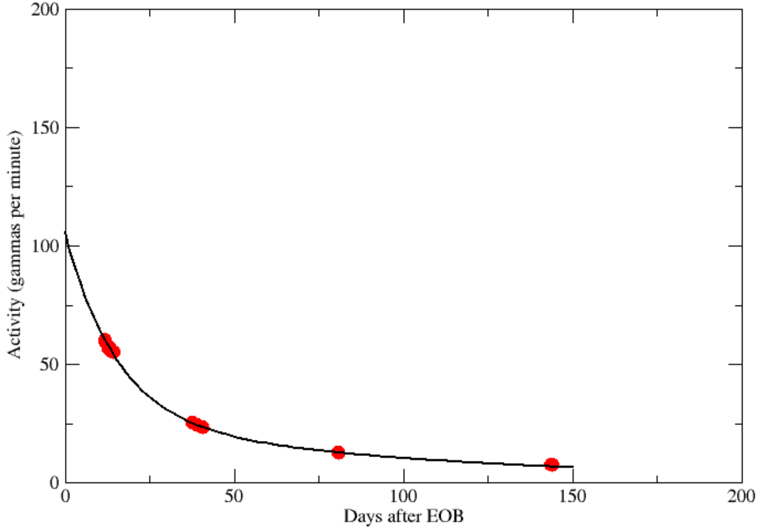


- The ^{192}Ir x-rays interferes with the ^{193m}Ir K_{a1} and K_{a2} x-rays.
- The ^{193m}Ir decays with a 10.5 day half-life and ^{192}Ir with a 73.8 day half-life allowing us to correct for this interference after ^{193m}Ir has decayed away.

Status of HFNG Experiment

- The 64.9 keV iridium Kα1 x-ray overlaps with the platinum Kα2 from ^{192}Ir.
- The count rate in the iridium foil was fit to a 10.53 day and a 73.831 day activity.
- The result is 79.1 ^{193m}Ir Kα1 x-rays per minute at EOB.
- The ^{192}Ir was assayed by the 316 keV gamma ray.
- The activity has been corrected for self-attenuation of the 65 keV x-ray and growth and decay during irradiation. But we are pending a correction for the neutron beam geometry.
- The ^{193m}Ir and ^{192}Ir activities at EOB were 638 and 8.95 Bq, respectively.
- Relative to ^{193}Ir(n,2n)^{192}Ir cross section the ^{193}Ir(n,n')^{193m}Ir cross section is 0.5 b.

64.9 keV x-ray decay curve



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  - Kathy Shield, UC Berkeley
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  - Gus Ridenour, GWU
- HFNG Experiment
  - Karl van Bibber, Lee Bernstein, Jon Morrell, UC Berkeley
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  - Jason Burke, LLNL
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  - NA-22